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Patent

## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicants : T. Xu et al.

Group Art Unit: 2337

Appl. No : 09/892,307

Examiner: T. D. Dang

Filed : June 26, 2001

For : PRODUCTION OF LIGHT OLEFINS FROM OXYGENATE USING  
FRAMEWORK GALLIUM-CONTAINING MEDIUM PORE  
MOLECULAR SIEVE

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AMENDMENT UNDER 37 CFR § 1.111

Commissioner for Patents  
Washington, D.C. 20231

Sir:

Responsive to the Office Action of October 7, 2002, reconsideration and withdrawal of the rejections of record are requested. Inasmuch as the three-month shortened statutory period was originally set in the Office Action to expire on January 7, 2003, this amendment is being filed in a timely manner.

IN THE CLAIMS:

Please amend claims 1 to 3, 6, 7 and 9 to 11 as follows (a clean copy of the claims appears directly below and a marked up copy appears in the Appendix):

1. (Amended) A method for converting oxygenates to olefins comprising contacting said oxygenates and an aromatics co-feed with a framework gallium-containing molecular sieve catalyst comprising pores having a size ranging from about 5.0 Angstroms to 7.0 Angstroms, under conversion conditions effective to produce olefins, said catalyst having a Si/Ga molar ratio ranging from 5 to 500.

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14. (Amended) The method of claim 1 wherein said catalyst is a zeolite bound zeolite having a bound framework Ga-containing zeolite having a binder of framework Ga-containing zeolite having a Si/Ga molar ratio ranging from 5 to  $\infty$ .

16. (Amended) The method of claim 1 wherein said catalyst comprises silicoaluminophosphate as said molecular sieve.

17. (Amended) A method for converting methanol and/or dimethyl ether to a product containing C<sub>2</sub> and C<sub>3</sub> olefins which comprises contacting a feed which contains methanol and/or dimethyl ether with a catalyst comprising a gallium-modified ZSM-5 porous crystalline material to produce the product containing the C<sub>2</sub> and C<sub>3</sub> olefins, said contacting being conducted in the presence of an aromatic compound under conversion conditions including a temperature of 350°C to 480°C and a methanol and/or dimethyl ether partial pressure in excess of 6.9 kPa, and said catalyst having a Si/Ga molar ratio ranging from 5 to 500.

18. (Amended) The method of claim 17 wherein said catalyst comprises zeolite-bound zeolite having a bound framework Ga-containing zeolite having a binder of framework Ga-containing zeolite having a Si/Ga molar ratio ranging from 5 to  $\infty$ .

Please add claim 22:

---22. A method for converting oxygenates to olefins comprising contacting said oxygenates and an aromatics co-feed with a framework gallium-containing molecular sieve catalyst comprising pores having a size ranging from about 5.0 Angstroms to about 7.0 Angstroms, under conversion conditions effective to produce olefins, said catalyst having a Si/Ga molar ratio ranging from 5 to 500 and wherein said conversion conditions comprise a temperature of from about 300° C to about 450° C.

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**REMARKS**

Reconsideration and withdrawal of the rejections of record are respectfully requested.

***Summary of Status of Amendments and Office Action***

Upon entry of the above amendment, claims 1, 14, 17 and 18 will have been amended and claim 22 will have been added. Accordingly, claims 1 to 19 and 22 will be pending in the application with claims 1, 17 and 22 being independent.

In the Office Action, claims 16-19 are rejected under 35 U.S.C. § 112, second paragraph; claims 1-12 are rejected under 35 U.S.C. § 102(b); claim 17 is rejected under 35 U.S.C. § 102(b)/103(a); claim 16 is rejected under 35 U.S.C. § 103(a); and claims 13-15, 18 and 19 are rejected under 35 U.S.C. § 103(a).

***Acknowledgement of the Information Disclosure Statement***

Applicants respectfully thank the Examiner for considering the documents submitted in the Information Disclosure Statement, filed March 25, 2002, and for returning a signed copy of the PTO Forms 1149.

***Amendments to the Claims and Newly Added Claim 22***

Claims 1 and 17 have been amended to recite that the catalyst has a Si/Ga molar ratio ranging from 5 to 500. Support for this amendment can be found in dependent claims 14 and 18. Consequently, Applicants submit that no new matter has been entered into claims 1 and 17.

Claims 14 and 18 have been amended to remove the recitation that the catalyst has a Si/Ga molar ratio ranging from 5 to 500, which recitation was added to claims 1 and 17.

Claim 16 has been amended to recite that that the catalyst comprises silicoaluminophosphate as the molecular sieve component. Support for this can be found at the sentence bridging pages 6 and 7 of the specification which states that "[s]ilicoaluminophosphates such as SAPO-34, and "non-zeolitic molecular sieves" (such

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as metal aluminophosphates) . . . can be treated with gallium and used in the present invention."

Newly added claim 22 recites a method for converting oxygenates to olefins, which comprises contacting said oxygenates and an aromatics co-feed with a framework gallium-containing molecular sieve catalyst comprising pores having a size ranging from about 5.0 Angstroms to about 7.0 Angstroms, under conversion conditions effective to produce olefins, said catalyst having a Si/Ga molar ratio ranging from 5 to 500 and wherein said conversion conditions comprise a temperature of from about 300°C to about 450°C. Support for the claim can be found in original claim 1. In addition, the Examiner should note that claim 22 further recites the preferred temperature range, which is discussed in the specification at page 18, lines 9 to 12.

***Response to the Examiner's Rejection of Claims 16-19.***

In the Office Action, the Examiner rejected claims 16-19 under 35 U.S.C. § 112, second paragraph, as being indefinite. The Examiner urges that claim 16 is unclear whether the "silicoaluminophosphate" is an additional component of the catalyst or is a molecular sieve containing Ga in the framework. The Examiner urges that claim 17 is indefinite because of the expression, "being capable of alkylation" because the Examiner considers it unclear whether the aromatic compound is alkylated during the process or not.

With respect to claim 16, Applicants direct the Examiner's attention to the specification at page 6, line 29 to page 7, line 2, which clearly indicates that silicoaluminophosphate is a material which can be treated with gallium, e.g., a molecular sieve. Thus, the recitation in claim 16 "said catalyst comprises silicoaluminophosphate as said molecular sieve" is sufficiently definite to meet the statutory standard.

With respect to claim 17, Applicants respectfully submit that the claim is not indefinite. Depending upon the process conditions, the aromatic compound either will be or will not be alkylated. However, to clarify, Applicants have amended claim 17 to recite "to produce the product containing the C<sub>2</sub> and C<sub>3</sub> olefins," thus more clearly stating that the C<sub>2</sub> and C<sub>3</sub> olefins are present in the resulting product, either reacted with the aromatic or unreacted with the aromatic or both.

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For the foregoing reasons, Applicants respectfully request that the Examiner withdraw the rejection.

***Response to the Rejection to Claims 1 to 12 Under 35 U.S.C. § 102(b)***

Claims 1 to 12 are rejected under 35 U.S.C. § 102(b) as being anticipated by U.S. Patent No. 4,638,106 to Pieters et al. (the Pieters patent).

In order for a reference to anticipate a claim, the reference must show each and every element recited by the claim. Claim 1 has been amended to recite that the catalyst has a Si/Ga molar ratio ranging from 5 to 500. The Pieters patent does not disclose a catalyst that has a Si/Ga molar ratio ranging from 5 to 500. Thus claim 1 is not anticipated by the Pieters patent.

Furthermore, claims 2-12 depend upon claim 1 and therefore contain all of the elements of claim 1. Thus if claim 1 is not anticipated by the Pieters patent, claims 2-12 are likewise not anticipated. Applicants respectfully submit that the Pieters patent no longer anticipates the subject matter of claims 1 to 12. Accordingly, the rejection is now moot. Applicants respectfully request that the Examiner withdraw the rejection.

***Response to the Rejection of Claim 17 Under 35 U.S.C. §§ 102/ 103(a)***

Claim 17 is rejected under 35 U.S.C. § 102(b) as being anticipated by or, in the alternative, under 35 U.S.C. § 103(a) as being obvious over the Pieters patent.

To anticipate a claim, the anticipating reference must show each and every element recited by the claim. Claim 17 has been amended to recite that the catalyst has a Si/Ga molar ratio ranging from 5 to 500. The Pieters patent does not disclose a catalyst that has a Si/Ga molar ratio ranging from 5 to 500. Thus, the Pieters patent does not anticipate claim 17. Accordingly, the rejection of claim 17 under 102(a) is now moot. Applicants respectfully request that the Examiner withdraw the rejection.

Insofar as obviousness under 35 U.S.C. § 103(a) is concerned, Applicants respectfully submit that the Examiner has not shown why or how one skilled in the art would have been motivated to modify the teachings of the Pieters patent with respect to its catalyst. As noted above, amended claim 17 recites that "said catalyst having a Si/Ga molar ratio ranging from 5 to 500" (i.e. *5:1 to 500:1*) and this recitation is not taught or

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suggested by the Pieters patent. In fact, Applicants submit that the Pieters patent teaches away from using such a Si/Ga molar ratio inasmuch as it specifies "an effective silica to gallia mole ratio, to impart . . . acidity thereto" (column 11, lines 38 to 41) it being well understood that a person knowledgeable of zeolite catalysis synthesis is that acidity is imported, in certain instances, by substituting trivalent atoms such as aluminum or gallium for tetravalent silicon atoms. In this regard, the Examiner's attention is directed to the Pieters patent, column 11, lines 38 to 43, teaching a silica to gallia ( $\text{SiO}_2/\text{Ga}_2\text{O}_3$ ) mole ratio of *from about 1:400 to about 1:100*, corresponding to a silicon to gallium mole ratio (Si/Ga), of *from about 1:200 to about 1:50*, which ratio is well outside Applicants' recited range of *5:1 to 500:1*.

For the foregoing reasons, Applicants respectfully request that the Examiner withdraw this rejection.

***Response to the Rejection of Claim 16 Under 35 U.S.C. § 103(a)***

Claim 16 is rejected under 35 U.S.C. § 103(a) as being unpatentable over the Pieters patent in view of U.S. Patent No. 4,861,938 to Lewis et al. (the Lewis patent).

In the Office Action, the Examiner states that the Pieters patent discloses a process as discussed in the above rejections. In those rejections, it is the Examiner's position that the Pieters patent discloses a conversion process including a step of contacting an oxygenate feed such as methanol and an aromatic co-feed such as xylenes in the presence of a zeolite such as ZSM-5 having gallium in its framework. The Examiner acknowledges that the Pieters patent does not disclose that silicoaluminophosphate is also included in the catalyst, but that the Lewis patent at the abstract, at column 19, lines 26 to 37 and column 20, lines 10 to 33, discloses using silicoaluminophosphate to catalyze the conversion of oxygenates to olefins. Thus, the Examiner urges that it would have been obvious to one having ordinary skill to have modified the process disclosed in the Pieters patent by using a mixture of the silicoaluminophosphates disclosed in the Lewis patent and the catalyst disclosed in the Pieters patent to catalyze the oxygenate conversion process, because it is obvious to combine two compositions each of which is taught by the prior art to be useful for the same purpose in order to form a third composition, citing In re Kerkhoven, 205 USPQ 1069 (CCPA 1980).

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Claim 16 depends from claim 1 and in the present amendment, Applicants have amended claim 1. Thus, Applicants will direct their remarks with respect to claim 16 as it depends from amended claim 1. Applicants respectfully submit that the Examiner has not made out a prima facie case of obviousness.

Contrary to the Examiner's position, the Pieters patent does not disclose, for the reasons as set forth above a method for converting oxygenates to olefins which comprises contacting said oxygenates and an aromatics co-feed with a framework gallium-containing molecular sieve catalyst comprising pores having a size ranging from about 5.0 Angstroms to 7.0 Angstroms, under conversion conditions effective to produce olefins, wherein the catalyst has a Si/Ga molar ratio ranging from 5 to 500. Nor does the Pieters patent disclose use of molecular sieve catalyst comprising silicoaluminophosphate as recited in dependent claim 16, which depends from claim 1. To overcome the acknowledged deficiencies of the Pieters patent, the Examiner relies upon the Lewis patent, which teaches the use of silicoaluminophosphate for promoting the reaction of methanol to form products. The Lewis patent, however, does not teach that an aromatics co-feed is also present in his method. Furthermore, the Lewis patent doesn't teach a catalyst that has a Si/Ga molar ratio ranging from 5 to 500.

Applicants submit that there is no motivation to combine the teachings of the Pieters patent et al. with those of the Lewis patent and that if the teachings of the Pieters patent et al. were combined with those of the Lewis patent, the combined teachings would not necessarily teach the method recited in claim 16, inasmuch as the skilled worker would not have been given any direction as to which teachings to combine together in order to arrive at the claimed invention. Further, Applicants submit that even if the teachings could be combined, the combined references would not teach a catalyst having a Si/Ga molar ratio ranging from 5 to 500.

Furthermore, the Kerkhoven decision concerns compositions, which contain two components, each component of which was known in the prior art to perform the same function, whereas the present claim 16 recites a method wherein the molecular sieve catalysts comprises silicoaluminophosphate. Although claim 16 does exclude the presence of an additional catalyst by its inclusion of the term "comprising," such an

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additional catalyst is not required. Thus, the facts in Kerkhoven are not similar to the facts here.

For the foregoing reasons, Applicants request that the Examiner withdraw this rejection.

***Response to the Rejection of Claims 13-15, 18 and 19 Under 35 U.S.C. § 103(a)***

Claims 13 to 15, 18 and 19 are rejected under 35 U.S.C. § 103(a) as being unpatentable to the Pieters patent in view of U.S. Patent No. 6,150,293 to Verduijn et al. (the Verduijn patent).

In the Office Action, the Examiner acknowledges that the Pieters patent does not disclose using a zeolite bound zeolite catalyst as called for by claims 13 to 15, 18 and 19. The Examiner thus relies upon the abstract in the Verduijn patent at column 5, lines 10 to 30 and column 9, section (u) for their teachings of using zeolite bound zeolite catalysts for converting oxygenates to olefins. Accordingly, the Examiner considers that the claimed method of claims 13 to 15, 18 and 19 would have been obvious.

Applicants respectfully traverse this rejection.

Applicants submit that there is no motivation to combine the teachings of the Pieters patent with those of the Verduijn patent, and that if the teachings of the Pieters patent were combined with those of the Verduijn patent, the combined teachings would not teach the method recited in claims 13 to 15, 18 and 19, inasmuch as the skilled worker is not given any direction as to which teachings to combine together in order to arrive at the claimed invention. For example, the specification of the Verduijn patent indicates that the disclosed catalysts are useful in a wide variety of processes and states that the catalyst would be useful in the conversion of oxygenates to hydrocarbons including olefins and aromatics. However, the Verduijn patent does not give any specifics as to the disclosed oxygenation process. For example, the Verduijn patent does not disclose the presence of an aromatic co-feed.

Assuming for the sake of argument that the Examiner has made out a prima facie case of obviousness, which he has not, Applicants have rebutted that case with unexpected results. Applicants respectfully direct the Examiner to the specification's data in Table 6 (at page 18). The data show that at higher temperatures (500° C and 550°C) as disclosed by the Verduijn patent, unwanted methane selectivity has increased, whereas at



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temperatures (400° C and 450°C) methane selectivity was significantly reduced. Applicants submit that a person skilled in the art would recognize from the data that temperatures lower than 400°C would also give similar results, i.e., a reduction in methane selectivity. Thus, Applicants submit that they have demonstrated unexpectedness in a narrower temperature range of 300° to 450°C which is within the temperature range of 275° to about 600°C disclosed in the Verduijn patent, column 9, lines 50 and 51 (section u).

Further, the Examiner's attention is directed to newly added claim 22, which recites a temperature range from about 300° C to about 450°C, which is within the temperature range of 275°C to 600°C. Applicants submit that the data show unexpected results for the narrower ranges now recited in claim 22.

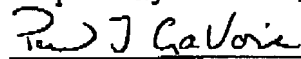
For the foregoing reasons, Applicants respectfully request that the Examiner withdraw the rejection.

#### CONCLUSION

For the reasons advanced above, Applicants respectfully submit that all pending claims patentably define Applicants' invention. Allowance of the application with an early mailing date of the Notice of Allowance and allowability is therefore respectfully requested.

Should the Examiner have any further comments or questions, the Examiner is invited to contact the undersigned at the below-listed telephone number.

Respectfully submitted,



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APPENDIX

## MARKED UP COPY OF CLAIMS

1. (Amended) A method for converting oxygenates to olefins comprising contacting said oxygenates and an aromatics co-feed with a framework gallium-containing molecular sieve catalyst comprising pores having a size ranging from about 5.0 Angstroms to 7.0 Angstroms, under conversion conditions effective to produce olefins, said catalyst having a Si/Ga molar ratio ranging from 5 to 500.

14. (Amended) The method of claim 1 wherein said catalyst is a zeolite bound zeolite having a bound framework Ga-containing zeolite having [a Si/Ga molar ratio ranging from 5 to 500 and] a binder of framework Ga-containing zeolite having a Si/Ga molar ratio ranging from 5 to  $\infty$ .

16. (Amended) The method of claim 1 wherein said catalyst comprises silicoaluminophosphate as said molecular sieve.

17. (Amended) A method for converting methanol and/or dimethyl ether to a product containing C<sub>2</sub> and C<sub>3</sub> olefins which comprises [the step of] contacting a feed which contains methanol and/or dimethyl ether with a catalyst comprising a gallium-modified ZSM-5 porous crystalline material to produce the product containing the C<sub>2</sub> and C<sub>3</sub> olefins, said contacting [step] being conducted in the presence of an aromatic compound under conversion conditions including a temperature of 350°C to 480°C and a methanol and/or dimethyl ether partial pressure in excess of 6.9kPa, and [the aromatic compound being capable of alkylation by the methanol and/or dimethyl ether under said conversion conditions] said catalyst having a Si/Ga molar ratio ranging from 5 to 500.

18. (Amended) The method of claim 17 wherein said catalyst comprises zeolite-bound zeolite having a bound framework Ga-containing zeolite having [a Si/Ga molar ratio ranging from 5 to 500 and] a binder of framework Ga-containing zeolite having a Si/Ga molar ratio ranging from 5 to  $\infty$ .